9. SHUFFLERS

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I. INTRODUCTION

A. Purpose of this Chapter

Shufflers were first conceived in 1969 as a method to assay uranium nondestructively. About 20 shufflers have been built in the US, and several others have been built in France and the United Kingdom. The purpose of this chapter is to introduce the technology, design, and operation of shufflers to readers who have little or no familiarity with this type of nondestructive assay system. A shuffler has a movable ^{252}Cf source that induces fission in fissile nuclei (^{235}U, ^{239}Pu) and is then moved away from the sample being assayed while a neutron counter measures delayed neutrons from fission. Shufflers are most often used to measure ^{235}U. The virtue of the shuffler technique is that the delayed neutron signal is proportional to the ^{252}Cf strength, yet the background is very low, because the source is shielded when delayed neutrons are counted.

For readers who want more technical detail than what is presented here, several published reports are cited as references. The Shuffler Application Guide, Ref. 1, is recommended as the first document to read, because it is the most comprehensive single reference to shuffler physics, design, calibration, and operation. The guide also includes an extensive bibliography of shuffler research at Los Alamos. Refs. 2–4 are also recommended. The first two discuss the detailed theory behind shufflers, with applications used as illustrations. The third report is concerned with the shuffler design most widely used to assay 200-liter drums (or smaller items) and discusses applications and results for assays of uranium and plutonium in a wide range of matrices.

B. The Motivation for Shufflers

1. History of Shufflers

In the 1960s, the Los Alamos safeguards group, under the direction of G. R. Keepin, began assaying fissile materials using delayed neutrons released after irradiation by neutrons from D-T reactions in a Van de Graaff accelerator. Measurements of delayed neutron parameters by Keepin and others formed a strong basis for applying delayed neutrons to uranium measurements.[5] When ^{252}Cf became available in the latter half of that decade, the advantages of using an intense isotopic source were quickly noted: reliable and predictable neutron emission rate, simple maintenance, and neutron energies too low to stimulate (n,2n) and (n,p) reactions. The main disadvantage was also clear: ^{252}Cf must be shielded at all times because it cannot be turned off. A D-T accelerator is likely to be less expensive than a ^{252}Cf source, but considering operating costs there is no significant monetary difference between the accelerator and isotopic sources.

Among the ^{252}Cf-based instruments proposed in 1969 was one in which a small capsule containing ^{252}Cf was “shuffled” pneumatically between storage and irradiation positions.[6] Fissions were induced in the fissile material and delayed neutrons counted after the ^{252}Cf source was returned to a shield. Figure 1 shows the “shuffling” process.

2. The Shuffler's Role Among Other NDA Instruments

Gamma-ray instruments perform excellent assays of uranium and plutonium samples of limited size. Gamma rays are more readily attenuated than neutrons, so even with good attenuation corrections there is a point where the attenuation is too large and assays are no longer possible. Before this point, plutonium samples emit enough neutrons from spontaneous fissions to make passive neutron counting feasible. Total neutron, coincidence neutron, and neutron multiplicity counters have evolved to provide accurate assays of plutonium. The shuffler technique does not compete with passive counters because spontaneous fission neutrons become a hindering background for a shuffler instead of a signal on which an assay is based; several exceptions will be noted later.

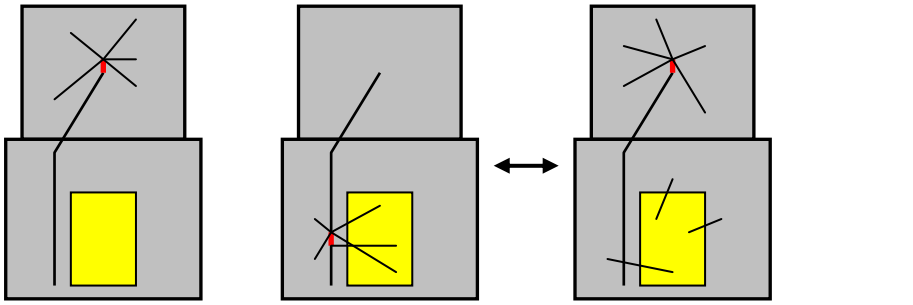

Fig. 1. The ^{252}Cf source alternates between the upper storage block and the lower assay chamber containing the object to be assayed (yellow). It is stored for the background count (a) and again after the irradiation (b) to count delayed neutrons (c). Steps (b) and (c) are repeated many times until the desired precision is reached. This "shuffling" of the source between two positions gives rise to the name of the instrument. For this illustration the source should be scanned along the side of the object during the irradiation (b) while the object rotates. Detector tubes surround the object to record the delayed neutrons (c).

However, uranium's passive neutron emission is far weaker than plutonium's and passive counters only work for large masses (> 1 kg) of ^{238}U. Active well coincidence counters (AWCC) were developed for uranium assay and their use overlaps some shuffler applications. An AWCC has AmLi sources above and below a can-sized container of uranium. Neutrons from the AmLi induce fissions and the fission neutrons are distinguished from the AmLi neutrons through their time correlations in time. The detected AmLi neutrons generate a high background that limits the precision (and sensitivity) of the AWCC.

An obvious niche for shufflers is in high-precision assays of bulk uranium where gamma-ray instruments cannot be used. Bulky items can be assayed with neutrons where gamma rays are strongly attenuated. The precision is better than an AWCC because the shuffler background during the count is very low. Since this initial application, the use of shufflers has expanded to other circumstances. For instance, a shuffler for bulk uranium can be a passive counter for bulk plutonium by keeping the ^{252}Cf source inside its shield while a passive count is taken. The shuffler's assay chamber can be designed to meet the criteria for passive counters, and one instrument then serves two roles.

3. Applications of Los Alamos Shufflers

This overview begins with an application for which shufflers are uniquely qualified: assaying large masses of dense uranium metal and oxide. Two variants of a shuffler for 200-liter drums

Fig. 2. The Uranium Scrap Shuffler had a hoist to raise and lower the heavy cans of scrap. A computer in an air-conditioned electronics rack was used to control the instrument and analyze the data.

are described, with applications to both large uranium masses and small waste quantities. Shufflers for unusual applications are also described. In these cases, high gamma-ray background rates or interference by neutrons from plutonium or curium make a shuffler necessary. Extensive details on all of these shufflers can be found in the bibliography.

a. Savannah River Uranium Scrap Shuffler

A shuffler (Figure 2) was built for the Savannah River Site (SRS) in 1978 to assay the mass of ^{235}U in cans of scrap metal from the billet extrusion process of making production fuel assemblies. This involved kilograms of uranium metal with enrichments from 50% to 80%. The spectrum-tailoring feature was retained to avoid interference from fissions in ^{238}U. Gamma rays could not be applied to these materials and the AWCC had just been developed. The only practical instrument was the shuffler, and its precision gave it an advantage over an AWCC.

b. Savannah River Billet Shuffler

The Billet Shuffler (Figure 3), installed at SRS in 1992, complemented the Uranium Scrap Shuffler. The Billet Shuffler determined the ^{235}U contents in billets prior to extrusion into fuel tubes. It was important to know the ^{235}U mass accurately to properly fuel the production reactors.

Fig. 3. Ed Adams opens the door to the assay chamber of the Billet Shuffler. The large cylinder in the foreground stores the ^{252}Cf source. Two simulated billets are on the cart to the right.

This shuffler had a precision of 0.25% (1σ) and an accuracy of 0.5% (1σ), even though the assay time (including background) was only 10 minutes. Such performance was possible because of the large ^{235}U loading (1.7 kg) and the billet's hollow core. A relatively small ^{252}Cf source (30 to 60 μg) was shuffled in and out of the center of the billet, making unusually efficient use of the irradiating neutrons. The excellent accuracy was made possible by the creation and careful characterization of calibration billets.

After the shutdown of the production reactors, the Billet Shuffler was used to measure uranium ingots prior to their shipment to Oak Ridge National Laboratory (ORNL). It assayed thousands of uranium ingots that previously would have become billets. The Billet Shuffler was then moved to a reactor building to help with decommissioning.

c. The "Standard" 200-liter-Drum Shuffler and Its Predecessors

There has an evolutionary progress in shufflers designed for 200-liter drums. In the late 1970s, a prototype shuffler for drums was created at Los Alamos by taking a passive neutron counter and adding a ^{252}Cf storage block and electronics. It was used to gain experience with 200-liter drum assays. Years later it was moved to Los Alamos's Chemistry and Metallurgy Research (CMR) Building to fill a temporary need and then was scraped. A unique feature of the shuffler was the shielding material in the walls of the assay chamber: water.

This shuffler was redesigned for Savannah River in 1984 to measure the uranium masses from fractions of a gram to hundreds of grams in a waste drum. Flux monitors were included to correct for matrix effects. These were two low-efficiency ^{3}He tubes that counted while the ^{252}Cf source irradiated a drum. One of the tubes was wrapped in cadmium so that moderators in a drum would affect the ratio of counts from these tubes. As a drum rotated on a turntable, the ^{252}Cf source scanned vertically to produce a distribution of delayed neutron precursors proportional to the distribution of uranium.

The shuffler was partially redesigned for the Portsmouth Gaseous Diffusion Plant to take advantage of improvements in electronics and ^{252}Cf motion control. Two of these shufflers (Figure 4) were delivered to Portsmouth in 1989 and 1991. Los Alamos commercialized this shuffler with Canberra Industries, which has since built two models for Los Alamos (Figure 5),

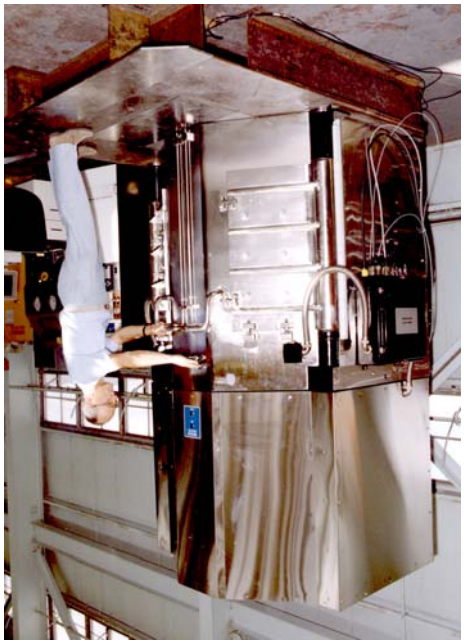

Fig. 4. Phil Rinard opens the doors of the Portsmouth Shuffler. Before shipping to Portsmouth, it was assembled and tested at Los Alamos on a riser. After installation, it sat flush with the floor. This design was commercialized through Canberra Industries.

Fig. 5. Sandra Hildner opens the doors of the shuffler for 200-liter drums at the Los Alamos Plutonium Facility to reveal a 200-liter drum. This shuffler is used to measure pyrochemical salt residues. These salts have very high ^{241}Am levels that produce large α,n rates that defeat passive coincidence methods. The shuffler can override this neutron background and achieve good measurement results.

one for Lawrence Livermore National Laboratory, and one for the Y-12 Plant at Oak Ridge. The applications of these shufflers have reached well beyond waste quantities to miscellaneous inventory items (grams to many kilograms of ^{235}U, ^{233}U, and ^{237}Np). The design in Figure 4 is now the “standard” 200-liter-drum shuffler.

d. The "Pass-Through" 200-liter-Drum and Boxed-Waste Shufflers

Savannah River requested two shufflers for 200-liter waste drums and boxed waste that could reside on materials accountability boundaries and decide, from an assay, whether a drum or box was allowed outside the boundary. A container inside the boundary entered the input door of the shuffler and was measured. If the uranium mass was below a preset limit, the output door opened and the container was now outside the boundary. Otherwise, the input door reopened and the container was kept within the boundary. This automated process avoided the costs of having to repeatedly open a boundary and use guards to check people in and out.

Fig. 6. The output side of the shuffler has a longer conveyer section to allow containers to be stored while new containers are assayed. Shown here are 200-liter drums, a HEPA filter, and a cardboard box

One of these shufflers had a conveyor system (Figure 6) that ran through the shuffler and moved the containers automatically. This shuffler was installed in 1993, not far from the Scrap and Billet shufflers. After the production reactors were shut down and fuel tube production ceased, it was adopted by Argonne National Laboratory at the Idaho National Engineering and Environmental Laboratory (INEEL).

e. Spent Naval Fuel Shuffler

The Spent Naval Fuel Shuffler (Figure 7) is one of the largest shufflers ever built. This shuffler, also known as the Fluorinel and Storage (FAST) Facility Shuffler, measured the uranium content in a spent-fuel assembly prior to reprocessing. Intense, fission-product gamma rays made it impossible to measure uranium gamma rays. In commercial reactor, low-enriched spent fuel, the buildup of ^{244}Cm creates an intense passive neutron rate that prevents a shuffler from detecting the much weaker delayed neutron signal from uranium (and also prevents passive counting for plutonium). But in highly enriched naval fuel, the ^{244}Cm buildup is much smaller, so this shuffler could assay for uranium directly. It required a 1- to 3-mg ^{252}Cf source, which is larger than usual for shufflers but not unusual in industrial applications of ^{252}Cf. The shuffler was built for the Idaho Chemical Processing Plant (ICPP), now known as the Idaho National Laboratory (INL).

Because the spent fuel had to remain within a hot cell, the shuffle body had also to stay there. Nevertheless, a large neutron shield was necessary to hold the ^{252}Cf source when it wasn't irradiating a fuel assembly and to protect workers who might enter the shuffler's general area. The ^{3}He tubes were shielded from fission-product gamma rays by 10 cm of lead. An assembly was moved vertically at a steady rate through the shuffler. The ^{252}Cf source performed its series of shuffles on the various segments of the fuel. In effect, the source scanned the fuel length, but the fuel did the vertical scanning. The shuffler became part of the reprocessing operations and

Fig. 7. George Eccleston and Tom Van Lyssel assemble the Spent Naval Fuel Shuffler for testing. A container is suspended above a small tube where assays are done on waste material inside the container. A larger tube to the left is for the fuel assemblies. A motor in the center ran an automated hoist. The motor at the bottom of the photograph is the stepping motor for the ^{252}Cf source.

proved to be very accurate. A second, smaller tube passing through the shuffler's body was provided to assay canisters of waste using the same ^{252}Cf source, but this capability was never applied.

f. Dounreay Reprocessing Solid-Waste Shuffler

The breeder reactor fuel reprocessing plant at Dounreay, Scotland, generates leached hulls and centrifuge bowls that may contain residual amounts of plutonium. Passive neutron counting is impossible in this case because ^{244}Cm, a prolific neutron emitter, is often present. Fission products and attenuation ruled out gamma-ray methods. The British installed a D-T generator to induce fissions in plutonium, but it proved a maintenance problem for a continuously operating plant. Los Alamos installed a shuffler at Dounreay in 1987 and it has been an integral part of the plant process ever since (Figure 8).

The shuffler takes a passive count. If no neutrons are found, there is no plutonium. If the passive count is positive, it may be caused by plutonium, curium, or both. An active assay is performed for the plutonium mass. A 3-mg ^{252}Cf source, when new, is used to override the worst ^{244}Cm case. Baskets of hulls or bowl parts are scanned past the irradiation position and the delayed neutron counts indicate the total plutonium mass. The shuffler's computer controls the hoist to ensure that the basket's position is properly synchronized with the ^{252}Cf source.

The computer, the electronics, and the stepping motors were upgraded in 1995.

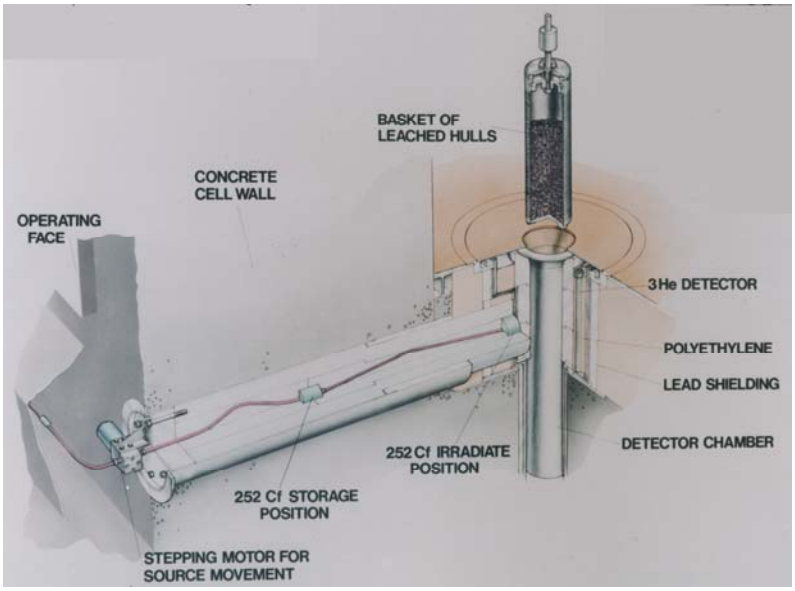

Fig. 8. The components of the Dounreay shuffler. A cylinder of polyethylene extends through the hot cell wall and the ^{252}Cf source is stored in the center of the wall. A basket of waste is lowered into the assay chamber while measurements are taken. The electromechanical components are in the corridor on the left edge of this drawing.

4. Applications of Shufflers in Europe

The nature and applications of shufflers in Europe are similar to those in the US. Initially, ^{252}Cf sources were moved pneumatically in shufflers in the UK and France, as was suggested in the first Los Alamos paper on the shuffler concept. It is a rapid way to move the source and uses technology familiar to reactor workers. However, scanning a large object during irradiation cannot be done and variations in the time used to move the source are large. Los Alamos shufflers use a stepping motor to move a strong, flexible, steel cable to which the source is attached. This technique is fast, reliable, and accurate. The source also can be easily moved at a slow speed to scan a large object (200-liter drum) during irradiation. The stepping motor and cable technique was adopted many years ago in both France and the UK. The French have even combined a more powerful motor with a large drum for winding the cable, resulting in even shorter times and longer distances to move the source.

Shufflers have been developed at Harwell in England for 200-liter drums, bags of waste, and mixed fissile materials. Liquid samples, dissolver tank residues, bags of waste, and 800-liter drums of leached hulls have been measured at the Cadarache Nuclear Center in France.

C. Shuffler Basics

The ideal measurement condition has a strong signal in the presence of a weak background. Uranium's passive neutron signals are weak and the passive gamma-ray signals are easily attenuated to the point where they cannot be detected. The AWCC generates a strong signal by inducing fissions and counting fission neutrons. However, the AmLi sources in the AWCC also produce a high background that limits the application of the AWCC to a ^{235}U mass greater than 10 g. Increasing the AmLi source strength beyond 10^5 n/s is counterproductive because the background rate also increases and the precision is harmed rather than helped. Furthermore, the size of the object measured by an AWCC is limited to a diameter and height of about 25 cm; the non-uniformity of the irradiating neutron flux becomes prohibitive beyond these dimensions.

The shuffler can generate the same or stronger signal as the AWCC, but with a much smaller background, even though it counts delayed neutrons that are less than 1% as numerous as prompt neutrons. The delayed neutron production rate is boosted by using a strong source (e.g., 10^{10} n/s) and the background rate is kept low by shielding the ^{252}Cf source during delayed neutron counting. The assay chamber can be relatively large (200-liter drum) without sacrificing uniformity of irradiation because the small ^{252}Cf capsule can be easily scanned and the drum rotated during the irradiation. For a detailed theoretical treatment of shufflers, see Refs. 1 and 2.

1. Delayed Neutrons

$$P(t) = \sum_{j=1}^{6} P_j(t) = \sum_{j=1}^{6} \beta_j \bar{\nu} e^{-\lambda_j t} \quad . \tag{1}$$

$P_j(t)$ is the population of the *jth* group of precursors, $\bar{\nu}$ is the average number of neutrons per fission, and β_j is the fraction of the fission neutrons that are delayed, so $\beta_j \bar{\nu}$ is the average of delayed neutrons produced per fission by group j. The decay constant for group j is λ_j. The parameters of the six groups vary among the uranium and plutonium isotopes (also thorium and neptunium) but are well known from measurements. These parameters are necessary to calculate required ^{252}Cf masses or expected count rates.

To gain a sense of their values, examine Table I for the parameters of ^{235}U and ^{239}Pu. The group half-lives are shown instead of the decay constants for quicker understanding of the behavior of delayed neutrons. (Uncertainties can be found in Ref. 2.) If one assay follows another, there should be a pause to allow the previous precursors to decay; 4 minutes is generally sufficient because the longest-lived precursors are not the most productive. The time decays of the precursors of these two important isotopes are shown in Figure 9; the amplitudes are per fission and reflect the different values of $\beta_j \bar{\nu}$. The two values of $\bar{\nu}$ in Table I are approximate because $\bar{\nu}$ actually has a slight dependence on the energy of the captured neutron.

TABLE I
DELAYED NEUTRON PARAMETERS FOR ^{235}U AND ^{239}Pu

	^{235}U. $\bar{\nu}$ = 2.43*		^{239}Pu. $\bar{\nu}$ = 2.88*	
Group j	$T_{1/2}$ (s)	$\beta_j \bar{\nu}$	$T_{1/2}$ (s)	$\beta_j \bar{\nu}$
1	55.72	0.00052	54.28	0.00021
2	22.72	0.00346	23.04	0.00182
3	6.22	0.00310	5.60	0.00129
4	2.30	0.00624	2.13	0.00199
5	0.610	0.00182	0.618	0.00052
6	0.230	0.00066	0.257	0.00027
sum	-------	0.01580	-------	0.00610

* These values are for thermal neutron fission. Actual values generally are slightly larger.

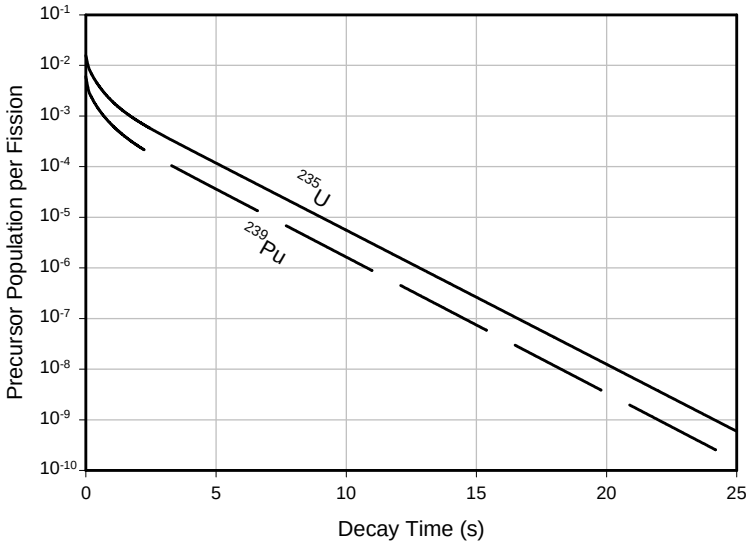

Fig. 9. After a fission, the average number of delayed neutron precursors present is shown as a function of time after the fission. The curve for an isotope is accurately expressed as a sum of six curves (one for each precursor group). Each group has an exponential decay but their sum isn't exponential until the short-lived precursors have decayed for about 5 s.

2. The Shuffler Principle

The principle behind a shuffler assay is illustrated in Figure 1. The general shape of the standard 200-liter-drum shuffler is used in this illustration. If an assay must be done in a specified time, optimum times of the various stages can be calculated to optimize the precision of the resulting count rate. For example, in the standard 200-liter-drum shuffler, the typical assay time is somewhat arbitrarily set at 1000 s. The background count uses the first 270 s of this time and the background is about 20-30 cps. Generally, 34 shuffles are performed, during each of which the irradiation is about 11.6-s long and the count time is about 7 s. The rest of the 1000 s (about 98 s) is spent moving the source and handling the intermediate data. The precision changes slowly as these times are modified, so the times are not critical. It is important that the times of all the steps are closely replicated during each of the shuffles; this is easily done by the computer and stepping motor to within a millisecond or so.

The user can easily modify any of these parameters to best match the current circumstances. If the assay time is doubled to 2000 s, the background time and shuffle number should also be doubled. The irradiation and count times for a given shuffle should not be changed because they are based on the nuclear parameters in Table I and the background rate. Should the background rate be much higher than 30 cps, the irradiation and count times should be reconsidered, as described in Ref. 2.

During a 7-s count most, but not all, of the delayed neutrons will be released (Figure 9). A small number will "carry over" into the next count time. Better use of the assay time is made by "sacrificing" some of the delayed neutrons and re-irradiating to replenish the precursor population. See Ref. 2 for more details.

3. Pertinent Properties of ^{252}Cf

The important property of ^{252}Cf for shuffler applications is its very high yield of spontaneous fission neutrons: 2.34×10^{12} n/s•g. This is 10^5 times greater than the yield of ^{244}Cm and 2×10^9 times greater than the yield of ^{240}Pu. It has the intensity of a small accelerator without the electronics and irregular variations in yield, plus a lower neutron energy. However, it cannot be turned off and it disappears with a half-life of 2.645 y.

For those who deal in curies, one gram of ^{252}Cf corresponds to 536.1 Ci. So 1 mCi of ^{252}Cf corresponds to a yield of 4.635×10^6 n/s, and 1 μCi gives 4635 n/s.

The half-life is short enough that a source must be oversized initially to have a useful lifespan. For a source to have a useful life of at least three years, it must be at least twice as large as necessary. Practical lifetimes are closer to 10 years, either because initial sizes are four times the minimum required, the precision is preserved by increasing the assay time, only materials with masses larger than the design minimum are measured, or some degradation of precision is acceptable.

Shufflers do not care that spontaneous fission neutrons are time correlated. The energy spectrum of spontaneous fission neutrons is very important. The usual expression for the spectrum is the Watt equation (Figure 10):

$$N(E) = e^{-E/a} \sinh(\sqrt{bE}), \qquad (2)$$

where E is the neutron energy in MeV and, for ^{252}Cf, a = 1.025 MeV and b = 2.926 MeV^{-1}. The average neutron energy is 2.3 MeV, but the most probable energy is 0.90 keV. The median energy is 3.27 MeV. This spectrum is used in the Monte Carlo calculations discussed later.

^{252}Cf is produced in the high-flux reactor at Oak Ridge. Table II presents the basic nuclear data used by the Radiochemical Engineering Development Center at Oak Ridge, where Cf is generated and sources are produced. John Bigelow and Joe Knauer kindly provided this information. For four of these six nuclides ($^{250, 251, 252, 254}$Cf) it is easy to calculate the emission rates given in Table II. The expression is:

$$\text{Emission rate} = \bar{\nu}\,(\text{SF Branching Fraction})\,[(\ln 2)/T_{1/2}]\,[6.02214 \times 10^{23} / (\text{At. Wt.})]. \qquad (3)$$

To get the emission rate in (n/g•s), the half-life must be in seconds. The spontaneous fission branching fraction is normally 1 minus the α-decay branching fraction. "At. Wt." is the atomic weight of the isotope.

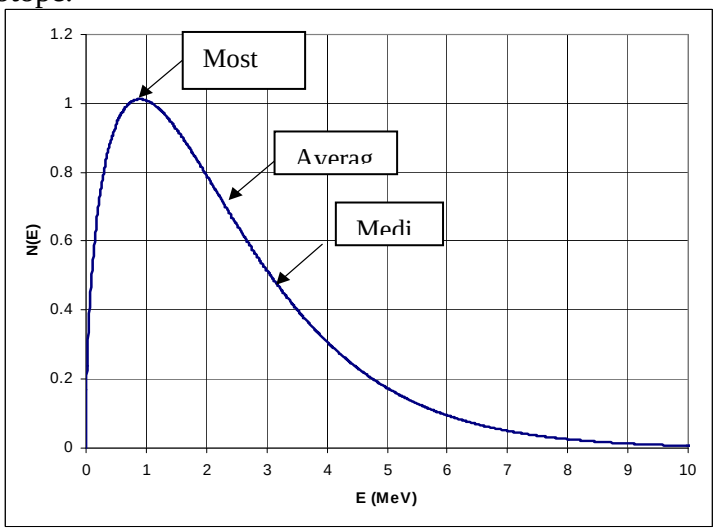

Fig. 10. The energy spectrum of ^{252}Cf neutrons (Watt equation). The average, most probable, and median energy are marked.

TABLE II
BASIC NUCLEAR DATA FOR Cf NUCLIDES

Nuclide	Half-Life ($T_{1/2}$)	α-Decay Branching Fraction	Spontaneous Fission (SF) Branching Fraction	$\bar{\nu}$ for SF	(α,n) Rate from Oxygen per Gram of ^{x}Cf	Total Neutron Emission Rate (n/g•s)
^{249}Cf	351 y	≈ 1.0	5.2×10^{-9}	3.4	≈ 3700	6.34×10^{3}
^{250}Cf	13.20 y	0.99921	0.00079	3.53	negligible	1.117×10^{10}
^{251}Cf	898 y	≈ 1.0	9.0×10^{-6}	3.7	negligible	1.955×10^{6}
^{252}Cf	2.645 y	0.96904	0.03096	3.768	negligible	2.314×10^{12}
^{253}Cf	17.81 d	0.0031	unknown	unknown	negligible	8.406×10^{4}
^{254}Cf	61.9 d	0.00299	0.99701	3.93	negligible	1.204×10^{15}

Fortunately, the worst uncertainties have no practical significance for shufflers because only two nuclides have important emission rates in a source: ^{250}Cf and ^{252}Cf. The production protocol is optimized for ^{252}Cf with its modest half-life, so the long-lived nuclides are not as abundant and the short-lived nuclides are gone before the source reaches a user. Figure 11 shows the relative neutron emission rates from ^{250}Cf and ^{252}Cf for three decades after creation. The initial relative

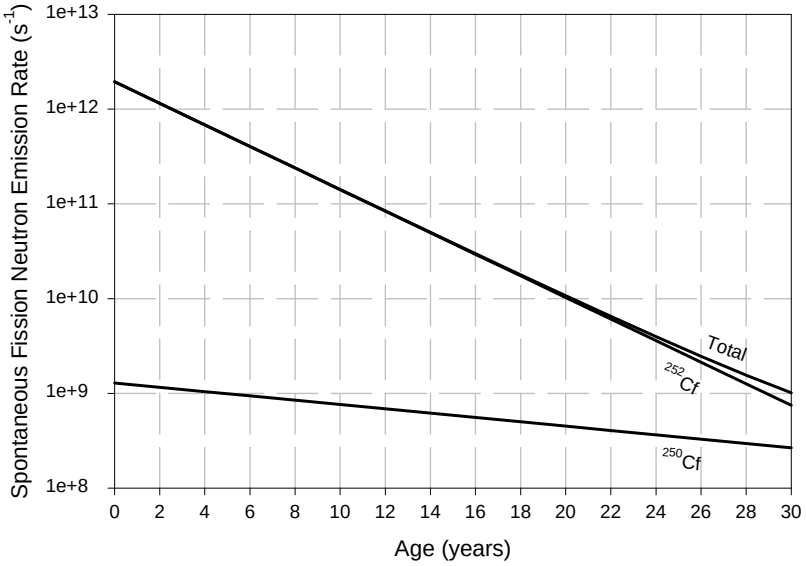

Fig. 11. Neutron emission rates as a function of time from one gram of Cf with the mixture of nuclides in Table III. The lowest curve is the rate from ^{250}Cf; the middle curve is the rate from ^{252}Cf; the upper curve is the sum of these two. The curves for ^{252}Cf and the sum are indistinguishable until an age of about 20 years.

abundances used are shown in Table III. The ^{250}Cf fraction of the neutron emission rate is shown in Figure 12.

TABLE III
INITIAL RELATIVE ABUNDANCES OF Cf NUCLIDES
This is a realistic example, but abundances vary among batches.

Nuclide	Relative Abundance (Fractional)
^{249}Cf	0.009424
^{250}Cf	0.115183
^{251}Cf	0.032461
^{252}Cf	0.837696
^{253}Cf	0.005236
^{254}Cf	0.000000

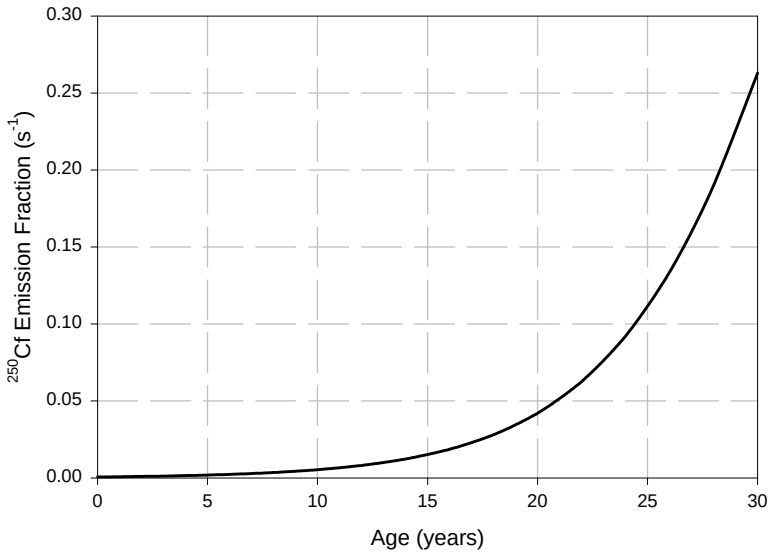

Fig. 12. The ^{250}Cf contribution to the total neutron emission rate from the Cf described in Table III increases with time because ^{252}Cf decays more quickly. It takes 13 years for ^{250}Cf to contribute 1%, but only an additional 8 years (21 years total) to reach 5%. The fraction grows to 25% in another 9 years (30 years total).

In practice, it is possible to ignore californium isotopes other than ^{252}Cf in shufflers because a source is not used for 20 years. An old shuffler source has other uses and its mixture of isotopes should be understood to avoid inaccurate results.

The neutron dose rate from 1 μg of ^{252}Cf at one meter without any shielding is 2.34 mrem/hr. The gamma rays contribute another 0.140 mrem/hr. The total dose rate is therefore 2.48 mrem/hr/μg at one meter with no shielding. The standard 200-liter-drum shuffler uses a 550-μg source when new, so the dose rate at one meter in air for such a source is 1.24 rem/hr. When loading a new source into a shuffler, the source is unshielded for about 15 s and nobody is closer than 2 meters. Under those conditions, the nearest person receives about 1.4 mrem. Doses measured during the loading process have been about 10 mrem, which includes the dose received from working around the shipping cask to prepare for the brief loading operation. The short time needed to insert a new source is the key to producing such a small dose to the handler and practice with a dummy source helps keep the time short.

An advantage of using ^{252}Cf for shufflers is the tiny mass and volume needed for a source with a large yield (e.g. 10^{10} n/s). All of the standard capsules overwhelm the size of the actual source; the capsules are made almost entirely of solid steel. The californium may be

electrodeposited on a small wire, which is cut in pieces to produce various neutron yields. Los Alamos uses standard, certified packages used for decades by Oak Ridge and Savannah River. The californium is double encapsulated in steel. The inner capsule has a small cavity for the wire, a diameter of about 0.5 cm, and a length of about 2.5 cm. This capsule goes inside an outer capsule, which has a diameter of about 0.75 cm and a length of about 3.7 cm. Both capsules are welded and have passed extensive durability tests under conditions much worse than in a shuffler.

One end of the outer capsule has a threaded nub. A steel coupler screws onto this nub and also onto a flexible steel cable with a wire wound around the outside, which serves as a coarse thread. All threads and set screws are cemented with LokTite to ensure that they will not come undone. The many sources attached in this manner have never accidentally detached from the cable, despite the immense number of shuffles of the sources.[7]

4. Factors That Complicate Assays

While it is easy to place an object in a shuffler and record the delayed neutron count rate a few minutes later, converting the count rate to an accurate mass of ^{235}U (or other fissile isotope) requires careful preparation. Calibration is important and not always simple.

When measuring ^{235}U, there is always some ^{238}U present. The probability of ^{238}U fissioning is essentially zero unless the energy of the absorbed neutron is more than 1 MeV. The ^{252}Cf energy spectrum of Fig. 10 makes it clear that, unless modified, there will be fissions in ^{238}U. Actually, some ^{238}U fissions occur regardless of the energy spectrum of the irradiating source because fission neutrons from ^{235}U have an energy spectrum not too different from ^{252}Cf. Some relief comes from the lower probability that a high-energy neutron will induce a fission in ^{238}U, but this is less than a factor of two. With high-enriched uranium there is more relief from the small quantity of ^{238}U (10% or less). On the other hand, the average number of delayed neutrons from a fission of ^{238}U is 2.6 times that of ^{235}U. The ^{238}U complication can be nearly eliminated with "spectrum tailoring," reducing the energies of the ^{252}Cf neutrons below 1 MeV before they reach the uranium. This is done by placing additional steel between the ^{252}Cf source and the measured sample.

While fissions in ^{238}U require high-energy neutrons, fissions in ^{235}U are more likely as the neutron energy decreases. The various matrices that might be put into the standard 200-liter-drum shuffler have a wide range of effects[4] that have been dealt with in various ways. Within such a large volume as a 200-liter drum, the same mass of ^{235}U can produce different count rates depending on its distribution throughout a moderating matrix, so a method of determining the distribution and correcting for it has been developed and implemented.[8]

II. Shuffler Performance

The performance of any NDA instrument is expressed by a set of parameters such as:

- Precision or Reproducibility. These words are synonyms, but only "precision" will be used. If a measurement is repeated many times, the precision is the standard deviation of the results. It may be expressed in the same units as the measurement, or as percent of the average of the measurements. In the latter case, it is called a relative precision. The standard deviation is often expressed in units of the σ of normal (Gaussian) distributions, where $\pm 1\sigma$ gives the range of values in which 68.3% of the measured values should fall. The item measured may be left untouched during this process or removed and replaced

between measurements to include handling effects. The time span of the measurements may be short (less than a day) or long (many days or years) to exhibit stability over time.

- Accuracy. This is a comparison of the measurement result and the best estimate of the true value. The difference is usually expressed as a percent of the true value; a smaller percent means better accuracy. Sometimes "bias" is used in this same sense. Sources of inaccuracy are sorted into systematic and random. The most notorious random causes are inherently beyond full control. Examples are the randomness in radioactive decay and backgrounds from cosmic rays. Cosmic rays can be mitigated with shielding and other techniques, but because it is impractical to work in deep mines, the cosmic-ray problem is almost always present. Systematic inaccuracies are caused by incomplete knowledge of the instrument, its calibration standards, or the materials being measured. Whatever errors exist in the calibration standards are passed along to the assay results.
- Sensitivity or Minimum Detectable Mass. These two terms are synonyms here, but whichever is used requires some supplementary information before it has meaning. The usual way to express sensitivity, the smallest mass that can be said to be measured with a stated certainty, is to relate the signal and noise "strengths" and define when the signal is just distinguishable from fluctuations in the noise. In shufflers, the signal is the delayed neutrons, while the noise is the background neutrons. A common practice is to define the sensitivity as the mass of fissile material that generates a delayed neutron count rate that is 3 times the 1σ error (random and systematic) of the background rate.
- Assay Time and Throughput. These are related, but not identical, concepts. The assay time is the time it takes to perform a complete measurement. Throughput is how many measurements can be completed in a given time. Throughput includes the time needed to change samples and enter information into the computer. The assay time may be 1000 s (16.67 min.) but the throughput might be three items an hour. Assay time directly affects precision and sensitivity, and it affects accuracy through precision.

With terminology defined, each performance characteristic will be examined for shufflers.

A. Precision

Precision is a shuffler strength. An intense ^{252}Cf source can be used to generate a large count rate of delayed neutrons and yet the background is low because the source is shielded while delayed neutrons are counted. The precision also depends on the mass and type of fissile isotope present. The probabilities of fissioning ^{235}U or ^{239}Pu are quite similar over a wide range of neutron energies, but ^{235}U produces about 2.6 times as many delayed neutrons per fission while spontaneous Pu fissions raise the background rate, affecting the precision. Everything else being equal, the precision with ^{235}U is better than with ^{239}Pu. This is also true when ^{235}U is compared with ^{233}U, ^{238}U, and ^{237}Np, just to name a few more isotopes.

The shuffler hardware affects the precision. Most of the hardware is fixed. However, the assay chamber is usually lined with cadmium to keep very low energy neutrons out of the chamber; this liner may be removed in just a few minutes. With everything else held the same, the precision can be greatly improved by not using a cadmium liner. The low-energy neutrons emerging from the moderating walls have high probabilities of inducing fissions and the count rate is greatly boosted. This can be effective in some circumstances, but it is generally a poor assay technique. The goal of an assay is not to get the best precision, but to get the best possible accuracy. If low-energy neutrons are allowed to dominate the fissions, the correlation between

count rate and mass becomes more complex as the mass. The self-shielding effect is accentuated and accuracy suffers. Table IV lists ways to improve precision, with cautionary notes.

B. Accuracy

Accuracy requires precision, but precision does not ensure accuracy. If the delayed neutron count rate is poorly known, the best calibration curve in the world cannot produce an accurate result. But a count rate with fantastic precision is meaningless without an accurate calibration curve. Accuracy is a relative term and depends on the use to be made of the result. A verification measurement checks a declared value, but is not as good as the declared value and does not replace it; it does not demand great accuracy. Accountability measurements should have the best possible accuracy and may replace declared values. For waste quantities, a poor accuracy may be tolerable because a large error in a small mass is still a small mass; a 100% error in 1 g is only 1 g. However, improving the accuracy becomes important when the measured mass plus some multiple of its uncertainty bumps into a regulatory limit. In this case, the waste may have to be placed in a category where handling and storage is more expensive.

TABLE IV - Improving Precision

To improve precision…	But be aware that…
Use a larger ^{252}Cf source.	More shielding is required and the size and weight of the shuffler grow exponentially.
Shorten the irradiation distance between the ^{252}Cf source and the fissile material.	If this produces a large inequality in the neutron flux for different places, the accuracy can suffer as the distribution of fissile material changes.
Use metal reflector behind ^{252}Cf to send more neutrons toward the fissile material.	This will never hurt the precision, but there may not be space for the 5-10 cm of iron (or other dense metal).
Surround the ^{252}Cf with metal to "tailor" the neutron energy spectrum toward lower energies.	Unwanted fissions in fertile isotopes (like ^{238}U) may be reduced, but self-shielding effects will be greater and the source will have to be somewhat stronger.
Use thermal and epithermal neutrons.	Self-shielding is a much larger problem even though count rates may increase.
Measure larger fissile masses.	The choice of masses is usually beyond the user's control. Self-shielding and multiplication effects grow, sometimes balancing each other nicely, but usually not.
Use longer assay times.	Throughput will decrease.
Use low background rates.	Poorly designed shielding may increase cosmic-ray interactions and hence the background rate. A larger storage block for the ^{252}Cf source reduces the background rate, but is the reduction commensurate with the increase in cost and improvement in performance?
Use more detector tubes or tubes with better sensitivity.	The procurement and fabrication costs increase.

The following shufflers made for Savannah River were specified to have exceptional accuracies because kilograms of uranium were involved: the Scrap Shuffler (0.3% for chips; 1.5% for floor sweepings), the Product Oxide shufflers (0.36%), and the Billet Shuffler (0.5%). On the other hand, the Liquid Raffinate Shuffler was specified by the Westinghouse Idaho Nuclear Co. to have an accuracy of 10% because it was more important to complete an assay in 100 s with a concentration of only 0.034 g/l (0.07 g-^{235}U in the 2-liter assay chamber).

Because a shuffler's precision is usually very good, accuracy is primarily set by the quality of the calibration. For the excellent accuracies of the three Savannah River shufflers mentioned above, the calibrations were extensive. Standards were carefully prepared and sampled for chemical analysis. For the Billet Shuffler, the billets were fabricated to small tolerances, so variations among billets were small except for the ^{235}U enrichment. The metal pieces for the Scrap Shuffler had irregular geometries, but this was not a major point. The Product Oxide shufflers had to work with oxides whose densities could vary, but with careful handling the variation was slight. With the development of very accurate calculational techniques (see Reference 3), the ability to calculate calibrations is possible and has been applied by Livermore and Los Alamos.

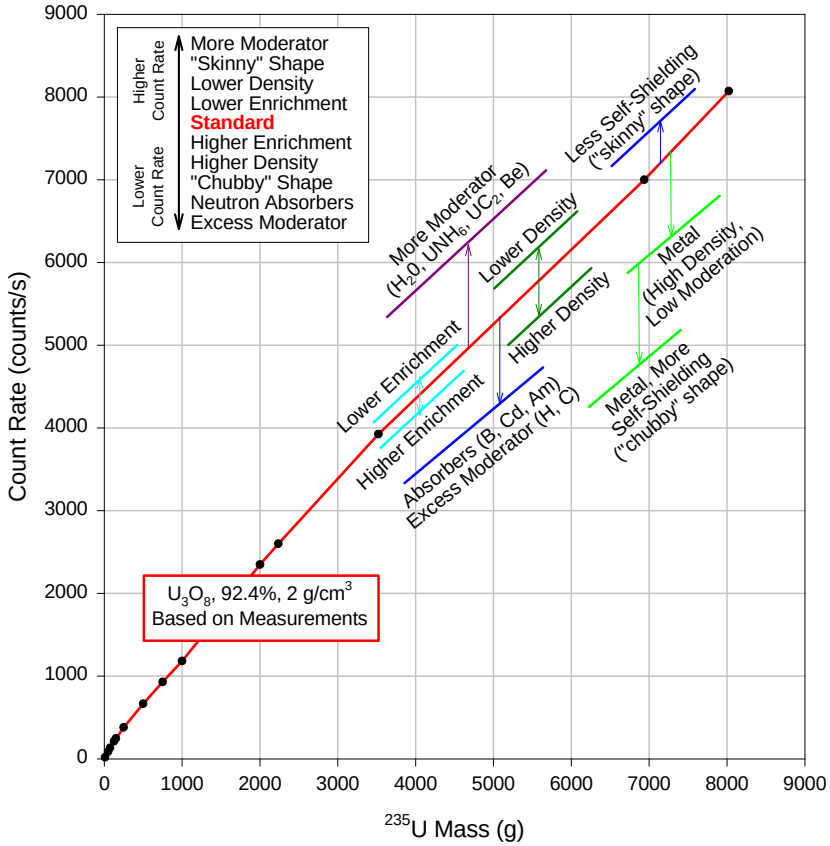

Fig. 13. When the material being measured has characteristics different from the calibration standards, the accuracy can be adversely affected. In this example, the calibration shown by the red line is for U_3O_8 with 92.4% ^{235}U and 4 g/cm^3. Potential changes are shown and the qualitative effect of each is indicated (exaggerated for clarity).

Standards based on powders (e.g. U_3O_8) require special attention because powder density changes with handling or even while sitting on a shelf. The powder density can be changed by as

much as 20% by shaking or tapping a can.[9] This changes the self-shielding of the material and, therefore, the count rate. A handling procedure should be established and adhered to strictly to make sure the calibration curve is appropriate for each measurement.

Figure 13 shows qualitatively how material variations can affect the accuracy of the calibration curve. Figures 14 and 15 are examples of measured and calculated effects of changes in chemical and physical form.

There are several ways to extend a calibration and minimize the number of standards. Calculational techniques are particularly useful in this context. If all the details of the model are correctly described, the accuracy of the result is as good as measured values. Extension of traditional standards is also possible. For example, a 200-liter drum of waste paper has a large amount of hydrogen that affects the count rate. A calibration done with one amount of paper has an accuracy problem for another drum with a paper density that differs by, say, 20%. You could have another calibration curve to handle this drum, but how would you know which calibration to use? The use of flux monitors and polyethylene sleeves and the measurement of fissile material distribution to address this problem are discussed later. Table V lists some ways to improve accuracy.

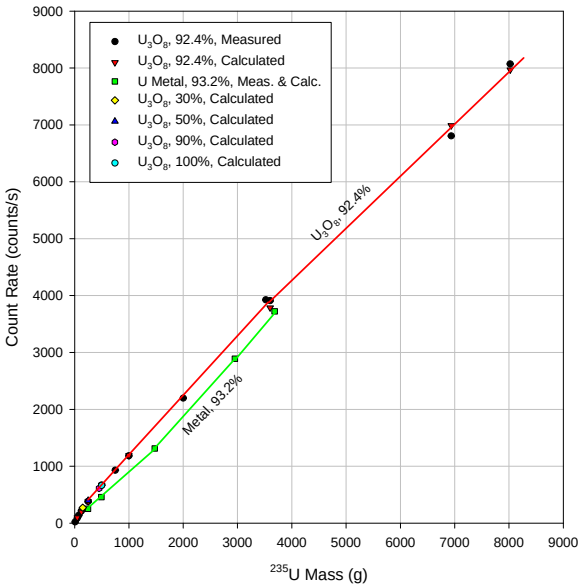

Fig. 14. The curve in red is the measured calibration for dry U_3O_8, 92.4% enriched. The effect of deviations from this have been calculated and are shown for comparison. Using metal disks (6-cm diameter) greatly reduces the count rate for the same mass of ^{235}U because of self-shielding from the higher density (19 instead of 4 g/cm^3). Enrichment effects are better seen in Fig. 15

TABLE V - Improving Accuracy

To improve accuracy…	But be aware that…
Improve the precision.	See potential problems given in Table IV.
Use calibration standards that match what you wish to measure and are known to at least the accuracy needed for the assays.	These can be expensive and may even be virtually impossible to build.
Control packaging and the matrix materials to match your standards.	This may be out of your control, despite your best attempts.
Handle containers of powders (e.g., U_3O_8) in a consistent manner to avoid changing the densities.	Some trial and error with handling techniques may be necessary to discover what works. Changes in density during storage may occur. Training users will be needed.
Scan larger objects with the ^{252}Cf source during the irradiations to create a distribution of precursors matching the distribution of fissile material.	Some trial and error with scanning techniques may be necessary to find the best one.
Use flux monitors to measure changes in hydrogen (moderator) density.	A measurement program with different moderator densities is needed to calibrate the flux monitor rates.
Use count rates in individual detector banks to find the distribution of fissile material within a moderating matrix.	A measurement campaign is needed to correlate the count rates with the different possible distributions.

C. Sensitivity or Minimum Detectable Mass

What is the smallest fissile mass that can be measured with a high probability (e.g. 95%) such that the signal is not a statistical fluctuation of the noise? That is one way to define sensitivity or, more descriptively, the minimum detectable mass. A shuffler cannot distinguish a delayed neutron from neutrons caused by cosmic-ray interactions or from the stored ^{252}Cf source.

The starting point for calculating the sensitivity is to express it in mathematical terms as the ratio of the delayed neutron counts, D, to the total uncertainty in the background counts, σ_B, This ratio is usually set to 3, to express the requirement that the signal be larger than the fluctuations in the noise. If $D/\sigma_B = 3$, it is sometimes said that the signal is 3σ above background. When the minimum detectable mass, m_s, is present, the delayed neutron counts have the special value of D_s. The two are related through the calibration function, after converting the counts into a rate over a count time T_D.

$$\frac{D_s}{T_D} = f(m_s) \qquad (4)$$

The uncertainty in a shuffler's delayed neutron count has a simple expression.[2]

$$\sigma_D = \sqrt{D + B\frac{T_D}{T_B} + B\left(\frac{T_D}{T_B}\right)^2} \quad , \qquad (5)$$

where

D = delayed neutron counts (not rate),
B = background counts (not rate),
$T_{D,B}$ = count time for delayed neutrons and background

As for all instruments, the sensitivity is limited by the background rate. Shuffler background rates are generally 10-40 cps, depending on the design and working environment. The definition of sensitivity is expressed with D_s/σ_B set usually to 3. The minimum necessary number of delayed neutron counts, D_s, follows from taking the ratio of Eqs. (4) and (5).

$$\frac{D_s}{\sigma_B} = \frac{T_D f(m_s)}{\sqrt{T_D f(m_s) + B\frac{T_D}{T_B} + B\left(\frac{T_D}{T_B}\right)^2}}. \qquad (6)$$

The minimum detectable mass m_s is found by solving this equation. If the calibration is a straight line through the origin (as for waste), this is easy to do algebraically.[3] For the most complex calibrations Eq. (6) can be solved by numerical methods (such as the Newton-Raphson method).

D. Choosing the Assay Time

In any NDA instrument, the assay time is usually a compromise that best meets conflicting specifications. The comparison in Table VI shows more advantages of using long assay times, but in practice rather short assay times (10 to 16 min.) are generally used because throughput is the main driver.

TABLE VI - Assay Times

Advantages of Long Assay Times	Advantages of Short Assay Times
Better precision.	Better throughput.
Better sensitivity.	Better immunity to changing conditions (e.g., background)
Can use smaller ^{252}Cf for a given precision, reducing cost and size.	
Can use smaller detection efficiencies for a given precision, reducing cost.	

Equation (6) is a good way to estimate the assay time needed to reach a measurement goal. The background and delayed neutron count rate must be known as well as possible. The latter depends on the ^{252}Cf mass, so there are many linked factors. As a rule of thumb, about a quarter of the assay time is spent on a background count. If the background rate is much different than the 10-40 cps mentioned earlier, a new study should be made of the fraction of total time spent on background counting. It is possible to do one long background count and use it for all assays of the day (or more), if it is clear that the background rate will not change. This would be the

most efficient use of all the time available, but it is rarely done because it is not certain that the background rate is constant. The background rate was *not* measured with each assay for the Liquid Raffinate Shuffler because the assay time had to be a short as possible (100 s) to ensure catching a high ^{235}U concentration as quickly as possible. A background count was taken periodically and used for many succeeding assays.

Los Alamos shufflers have always used an assay time set before the assay begins, expressed through the background time and the various parameters defining the shuffles. An alternative is to terminate the assay when a desired relative precision has been reached, up to some maximum number of shuffles. The average count rate then should be adjusted for the difference in the number of shuffles just used and the number used during calibration. This is another source of inaccuracy that is easily avoided by using a fixed assay time.

III. CREATING A SHUFFLER

A. Basic components

1. Hardware

A shuffler design centers on the assay chamber, which must be large enough to hold the biggest container, but no bigger than necessary. A snug fit between the assay chamber and the objects to be assayed is ideal, but even a small can may be quite adequately assayed in the 200-liter-drum shuffler despite the unequal sizes. Most containers should be rotated to make the irradiation more uniform throughout their volumes.

The ^{252}Cf source needs a storage block to reduce the dose rate from the source to the level specified by the user. In Los Alamos shufflers, the ^{252}Cf source is moved with a stepping motor that drives a cable attached to the source; positioning is quick and accurate. An alternative used in older European shufflers is to drive the source pneumatically. This is fast, but it can place the source at only two positions and cannot be used for scanning. Pneumatic methods have been dropped in favor of stepping motors.

The assay chamber is surrounded by neutron detectors, which are ^{3}He tubes with 2.5 cm diameters and pressures of 4 atm. For higher detection efficiency, the gas pressure can be as much as 10 atm. The tubes are electronically grouped and one signal cable receives the signals from several detectors, usually by connecting a set of detectors to one amplifier. The standard 200-liter-drum shuffler has six banks of detectors surrounding the side of the assay chamber, plus a bank above and a bank below the chamber. The amplifiers are now commercially available and built around the AmpTek 111 amplifier.

Flux monitors are also ^{3}He tubes, but are made less efficient to keep their interactions at a high but acceptable rate while the ^{252}Cf source is nearby. This inefficiency is accomplished by reducing the gas pressure, reducing the diameter, reducing the length, or some combination of these.

Figure 16 shows the electronic connections among the various shuffler components, with the coincidence counter as an optional feature. For a more detailed description of shuffler electronics, see the Application Guide.[1]

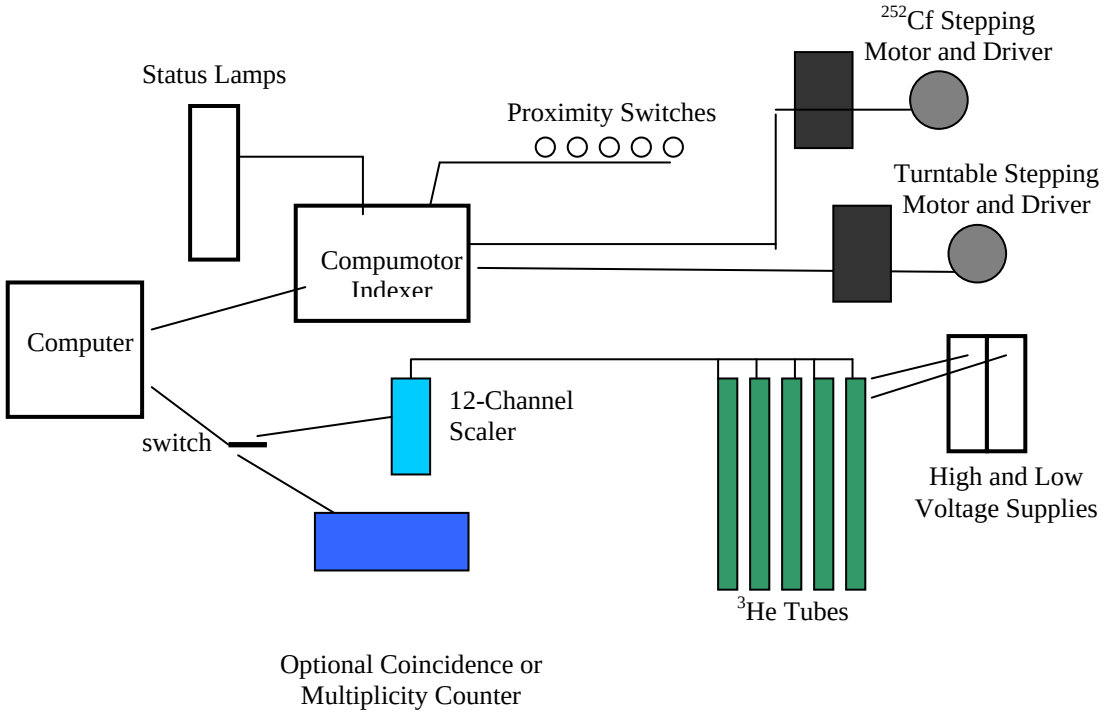

Fig. 16. The electrical connections among shuffler components. A serial data line from the computer normally goes to a 12-channel scaler, but for passive counting it is switched to a coincidence or multiplicity counter.

Shuffler computers have followed the evolution of computers themselves. Various DEC models and the FORTRAN language were used in the 1970s and into the 1980s. Then DOS-based machines and C were used from the late 1980s to the mid-1990s. Currently, the computers use a Windows variant and Visual C++. The basic operations and the results are independent of the platform chosen. The mathematical algorithms in the data analysis have been virtually unchanged throughout.

2. Software

The shuffler computer can run two entirely separate codes: the shuffler code and the INCC code for performing passive assays. Shuffler software is only covered briefly; the Application Guide[1] should be consulted for a more detailed description. Software controls the hardware and analyzes the neutron counts. Users can control software to the extent of setting important parameters and initiating measurement sequences, but the basic actions during a measurement are performed automatically by the computer. The shuffler software has several standard features, including:

- Perform assays, calibration measurements, precision measurements, bias checks, and standard checks.
- Perform safety checks to confirm that all proximity switches and lamps are working properly.
- Review archived results.
- Assign “general” parameters that are common to all measurements.
  - Nuclear constants: half-lives and delayed neutron yields for the six groups of precursor nuclei; the half-life of ^{252}Cf; and the reference date for the ^{252}Cf source.
  - Bias and precision check parameters.

- Assign “item-specific” parameters, one set for each container size and material type to be measured. Each type of item has a unique name given in the general parameters.
- Assay parameters: distances, speeds, accelerations, and pauses used for irradiation; length of the background count; and whether or not to rotate the turntable, and to use the flux monitors.
- Calibration parameters: Each item type has its own calibration parameters with variances and covariances. The standard expression provided is a third-order polynomial, but each item type can have more than one set of parameters for different ^{235}U enrichments.
- Diagnostics: This is a set of “primitive” actions that can be performed to check specific shuffler components (e.g. move source, store source, test door switch, test rotator, etc.)

Regardless of the operating system and programming language, shufflers do fundamentally the same things in the same way. Most importantly, the data analysis code has been stable for more than a decade and the assay results are independent of the computer used.

3. Safety features

The most novel hazards associated with shufflers arise from the ^{252}Cf sources. The masses of these sources have ranged from 1 to 3000 μg, according to the measurement problem. A microgram is a small source, but if unshielded, the dose rate at one meter is 2.5 mrem/hr. The standard 200-liter-drum shuffler may have as much as 550 μg of ^{252}Cf that delivers 1100 mrem/hr at one meter if unshielded. Clearly, the shuffler must be designed to keep the source shielded at all times. A complete list of safety features is found in the Ref. 1.

- If the assay chamber door is not closed and securely latched, the Indexer cannot move the ^{252}Cf source from its store position.
- The amount of shielding needed is specific to the facility. Maximum dose rates on the surface of a shuffler are generally specified by the facility and have been lowered from the early days. The older limits were 5 mrem/hr in contact; more recently, 0.5 mrem/hr has been the standard.
- One way to reduce dose rates without increasing costs is to use a barrier that increases the distance from the shuffler’s surface to people. This may be a few inches or a few feet. For example, if the dose rate on the surface of a storage block 61 cm thick is 0.6 mrem/hr, a barrier of only 5 cm beyond the shuffler’s surface will have a dose of 0.5 mrem/hr on its surface.
- There is never any good reason to “hug” a shuffler during measurements.

B. Physics Design

The physics design defines parameters that will ensure the shuffler meets the performance specifications. It is a paper and computer study that is done before any design drawings are made. The gross features of the design are defined by the physics calculations. Actual shuffler design is treated only briefly here.

1. User Specifications

Ref. 1 provides an extensive list of questions that must be answered by the facility procuring the shuffler.

2. Minimum Delayed Neutron Count Rate

The next step is to calculate the minimum delayed neutron count rate that will meet the performance specifications. The set of specifications can be summarized by a sentence such as "Measure a 500-g sphere of 94% enriched U metal with 1% relative precision in 1000 s." The material has been specified along with the precision and assay time. In this section, only the relative precision and assay time are needed. The material description will be used in the next section.

If there are no background counts, the relative precision of a delayed neutron count D is $1/\sqrt{D}$. For 1% relative precision, D is 10,000 counts. The full 1000 s could be devoted to this count, so the minimum count rate needed is a leisurely 10 counts/s. The ^{252}Cf source needed for this case would be relatively small and will be calculated in a later section.

But having no background counts is not realistic. Cosmic-ray interactions can be reduced by shielding, and the neutrons leaking into the assay chamber from the ^{252}Cf source can be reduced by even more shielding. How much are you willing to pay in cost and space for a reduced background rate? No shuffler has ever taken any special measures to reduce cosmic-ray-induced neutrons, and typical background rates from cosmic rays are 15 cps to nearly zero. Leakage from the ^{252}Cf storage block may add another 15 cps or less. Background rates of 30 cps are not unusual, but they have also been lower. The minimum count rate needed is D_s/T_c, as given in Eq. (4). The relative precision is the ratio of the count uncertainty with the count, as given in Eq. (6).

For the example given above and a background rate of 30 cps, the relative precision is 1% after a 270-s background count and a 240-s delayed neutron count when D is 17,688 counts. This typical background rate has forced us to count about 77% more delayed neutrons to reach the same relative precision. This count (and the corresponding 73.7 cps) is not unusual, and is in fact routine for the shuffler to produce, but the right ^{252}Cf mass must be chosen.

3. The Minimum ^{252}Cf Mass, Detection Efficiency, and Assay Chamber Shape

The remaining shuffler design depends on the mass of the ^{252}Cf that is needed to generate the minimum count rate needed to meet the specifications. What is the smallest mass that provides the desired relative precision in the time allowed? The detection efficiency is also directly involved at this point and the shape of the assay chamber has a role in determining it. But neither of these can be known accurately without a detailed design. An iterative process can be applied. With some experience, only a single pass through the process may be needed.

Pick a plausible detection efficiency for the first iteration. Something close to 20% is common for shufflers with a single layer of comfortably spaced ^{3}He tubes in polyethylene, but as much as 60%, could be achieved with multiple layers of tubes. The energy spectrum for delayed neutrons should be used, not the spectrum for ^{252}Cf or other prompt neutrons.

Pick an approximate size and shape for the assay chamber. If the final shape will snugly fit around the container, a looser fit will lead to a larger answer for the minimum ^{252}Cf mass. If the object does not have axial symmetry, several orientations should be used in calculations to get an average.

Next, calculate the probability that a single neutron from the ^{252}Cf source will induce a fission in the specified material, using the provisional assay chamber. It is difficult to do this accurately without a Monte Carlo simulation because the probability of inducing a fission is a function of neutron energy. The Los Alamos Monte Carlo code MCNP has been successfully used to calculate the fission probabilities for shufflers at Los Alamos and in Europe. A proven code uses continuous analog techniques to track neutron interactions within the shuffler materials, so changes in energy of the neutrons are correctly taken into account.

Assume that a model of the shuffler and a 500-g sphere of uranium have been made and the calculated fission probability is 7.89×10^{-4} fission per ^{252}Cf neutron. This includes fissions caused by fission neutrons as well as ^{252}Cf neutrons. The average number of neutrons released from a fission is known for all the fissile materials of interest to safeguards (for ^{235}U this is about 2.43). The average ^{252}Cf neutron produces 1.92×10^{-3} fission neutrons, but only a small fraction of these neutrons are delayed. In the case of ^{235}U, that fraction is about 1.6%, so the average ^{252}Cf neutron produces 3.07×10^{-5} delayed neutrons. The detection efficiency can be calculated with MCNP for a specified array of ^{3}He tubes. For our example, assume the result is 24%.

Some delayed neutrons are released while the ^{252}Cf irradiation is still in progress. These cannot be distinguished from ^{252}Cf neutrons and do not contribute to the count. The fraction of neutrons that can be counted while the ^{252}Cf source is stored depends on the irradiation scheme. A 20% fraction is a representative value and will be assumed here. This fraction can be calculated from a set of times for the different stages of a shuffle and the number of shuffles.[2]

The average ^{252}Cf neutron has the following probability of leading to a delayed neutron (dn) count:

$$(3.07 \times 10^{-5} \text{ dn}/^{252}\text{Cf n})(0.24 \text{ reaction/dn})(0.20 \text{count/reaction}) = 1.47 \times 10^{-6} \text{ count/Cf n}.$$

If the ^{252}Cf emits one neutron per second, it will take almost a million seconds (12 days) to get one count. We need 73.7 cps, as calculated in the preceding section. One microgram of ^{252}Cf emits 2.34×10^{6} neutrons/s. So the minimum mass of ^{252}Cf needed to get 1% relative precision in 1000 s can be calculated as follows:

$$(m_{Cf})(2.34 \times 10^{6} \text{ n/s})(1.47 \times 10^{-6} \text{ count/Cf n}) = 73.7 \text{ counts/s},$$
$$m_{Cf} = 21.4 \ \mu\text{g}.$$

4. The Initial ^{252}Cf Mass

If you fabricate a shuffler and put in the minimum ^{252}Cf mass, it will meet the specifications today, but not next year. In 147 days the neutron yield from the californium will be 90% of the original yield. In 2.65 years the yield is cut in half. The initial ^{252}Cf mass must allow for the time span during which the specifications are to be met. If our uranium sphere example specifies a useful life of 6 years, then the mass after 6 years of decay will be 21.4 μg and the initial mass at time zero is 103 μg.

5. ^{252}Cf Shielding

Once a mass of ^{252}Cf is chosen, the overall dimensions of the new shuffler can be calculated. The storage block has one set of dimensions and the assay chamber has another. The assay

chamber must be large enough to hold the objects to be measured and have wall thickness to shield personnel from the ^{252}Cf radiations.

One microgram of ^{252}Cf produces a dose rate of 2.45 mrem/hr at one meter, if unshielded. Our 150-μg example would produce 368 mrem/hr at one meter, a rate that must be reduced to the facility's acceptable value with shielding. Assume that the facility wants no more than 0.5 mrem/hr in contact with the shuffler. Two decisions must be made: What material or materials will constitute the shields, and what are their thicknesses? The answers are different for the storage block and the assay chamber.[10]

a. Storage Block

For the 150 μg of ^{252}Cf example, what is the overall size of the shield that will give only 0.5 mrem/hr on contact? Our measured shielding data shows that we will have 0.5 mrem/hr on the surface of a 1.2-m-wide shield if the ^{252}Cf mass is 43 μg. In other words, 150 μg behind 61 cm of shielding will have a contact dose rate of 1.7 mrem/hr, not 0.5 mrem/hr. It is not clear how much larger the shield would have to be to meet the specifications because our data stops at a thickness of 66 cm. and the neutron dose rate has begun to flatten out with thickness.

b. Assay Chamber Walls

A storage block that has a width of 1.2 m means that the source positioned in the block's center is surrounded by 0.6 m of shielding. The simplest approach to the thickness of the walls of the assay chamber might seem to be to use 0.6 m. of the same shielding. But the detector tubes need to be in embedded in polyethylene that doesn't contain boron, and this polyethylene should extend behind the tubes for several inches. Furthermore, the storage block's small core of heavy metal is rarely used in the assay chamber, with the rare exception of taking shielding credit for the metals used in spectrum tailoring.

The inner geometry of the assay chamber is fixed independently of the shielding. Figure 17 shows a cross section of the standard 200-liter-drum shuffler that uses as much as 550 μg of ^{252}Cf. The assay chamber is hexagonal to approximate the circular shape of the drum. The guide tube for the ^{252}Cf source runs along a block of iron. It acts as a reflector to send 25% more neutrons toward the drum and does no important spectrum tailoring.

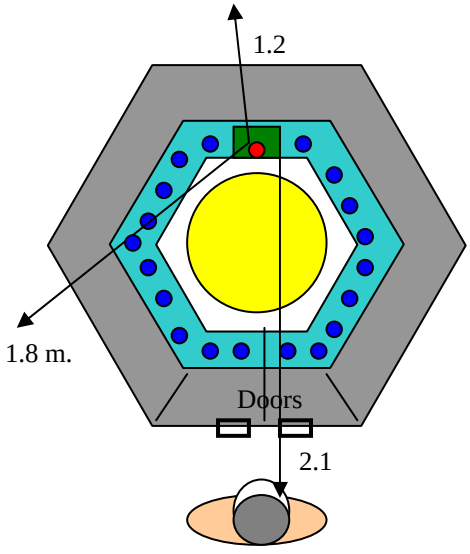

Fig. 17. A horizontal cross section of the standard shuffler for 200-liter drums is shown with an operator positioned in front of the doors. A drum is surrounded by detector tubes embedded in polyethylene, which is wrapped with an outer layer of borated polyethylene and 1.2 cm. of lead to form the shielding. Distances to the ^{252}Cf source and the amounts of shielding in the different directions are shown.

Detector tubes are embedded in polyethylene and surround the drum; there are more above and below the drum. The shielding beyond the detector region is composed of alternating layers of polyethylene and borated polyethylene, however, it is only about 36 cm thick.

6. Assay Chamber

The design of the assay chamber gets to the heart of a shuffler. The rest of the instrument supports the action within the assay chamber. The assay chamber generally has the first four components in the following list and may have any of the others.

- An empty chamber to receive the object to be measured.
- One or more doors that provide access to the chamber.
- Neutron detector tubes and amplifiers surrounding the chamber.
- A guide tube for the ^{252}Cf source and its flexible cable.
- An optional thin layer of cadmium around the inside wall of the chamber.
- An optional metallic reflector behind the source to send more ^{252}Cf neutrons toward the object.
- An optional spectrum tailoring set of materials.
- An optional turntable to rotate the object during the measurement.

7. Detector Tube Arrays

Detector tubes are usually embedded in banks of rectangular or semicircular pieces of polyethylene. A cross section of a rectangular bank is shown in Figure 18. Holes are drilled into the polyethylene at the tube position. A semicircular bank would have the same parameters.

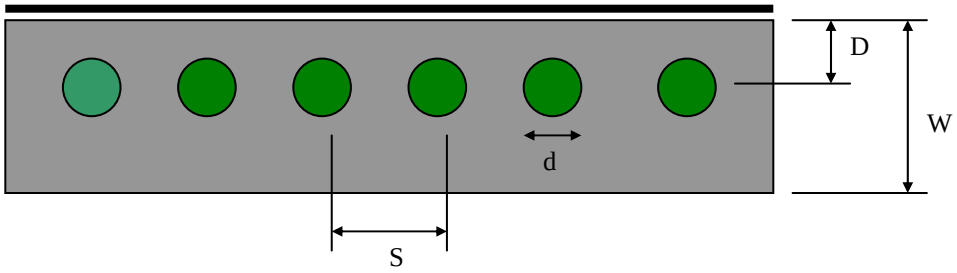

Fig. 18. A detector bank consists of ^{3}He tubes embedded in polyethylene. The heavy line at the top is an optional layer of cadmium and stainless steel lining the assay chamber. The assay chamber is located above the cadmium and steel liner. The important geometric parameters are indicated.

The width, *W*, of the bank is often 10 cm. because it works well and is a standard size for polyethylene. The tube diameter, *d*, is typically 2.5 cm, so the holes for the tubes are drilled slightly larger (e.g., 2.7 cm). The depth, *D*, between the tubes' centers and the side of the bank facing the fissile material affects the detection efficiency as a function of neutron energy, and the spacing, *S*, between tubes affects the detection efficiency for a given depth. For delayed neutrons, the depth, *D*, is smaller than the depth used in passive neutron counters for spontaneous fission neutrons. A general-purpose depth for shufflers is 3.8 cm. A spacing, *S*, less than 5 cm is counterproductive because a tube absorbs low-energy neutrons from its immediate vicinity and the tubes compete for these neutrons when they are too close together. They can be spaced as far

apart as desired if the detection efficiency needed is still met. Monte Carlo calculations are useful in designing the array of tubes needed to meet the shuffler's specifications

8. ^{252}Cf Motion Requirements

Delayed neutrons are produced during the irradiation, but cannot be counted against the background from the ^{252}Cf source. During the irradiation, their emission rate grows because many precursors have lifetimes longer than the irradiation time. When the irradiation ends, the emission rate of delayed neutrons begins to diminish rapidly. After an 11-s irradiation of ^{235}U, most of the delayed neutrons are released in the next 20 s. Obviously, you will count a larger fraction of these delayed neutrons if the ^{252}Cf source is removed quickly. This defines the most important feature of whatever is to move the source within the shuffler. It is less crucial to move the source into the irradiation position quickly, but a rapid motion here makes the mathematical expressions of a shuffler actions more accurate.

How quickly should the source move from the assay chamber back to the storage block? Some early Los Alamos shufflers used stepping motors tuned to run as fast as possible, and these sources moved about 1.5 m in about 0.33 s. But the tuning process was time-consuming and unique for a given set of equipment, and aging effects would require periodic retuning. It was decided to give up 10% of the delayed neutrons and use a slower, but very reliable, stepping (or servo) motor system from Compumotor that needs no maintenance. A move of 1.5 m is now generally done in about 0.75 s. To make up for the 10% drop in counts, the mass of the ^{252}Cf source can be increased by 10% or the assay time can be increased by about 10%. This sacrifice is well worth the elimination of the maintenance problems.

IV. CALIBRATION PROCEDURES

The shuffler measures a count rate of delayed neutrons and we must know how to convert that to fissile mass (^{235}U, ^{239}Pu, etc.). We can tell the shuffler's computer how to do the mathematics for us, but we have to figure it out ourselves first.

The best way to determine the calibration is to measure physical standards in the shuffler that accurately represent the items to be measured. The standards ideally match the items to be measured in all relevant respects (materials, size, shape, density, mass, isotopics, packaging, moisture content, and matrices). Standards usually have to be certified, meaning that the procedure used to fabricate the standards is accepted by regulatory agencies and a paper trail follows the standard to show how the procedure was followed. Sometimes such ideal standards exist, sometimes they can be made, but many times they do not exist and never will be made because of variations among the items.

A. Matrix Issues

The matrix is all the material in the item except the fissile material you want to measure. If you want to measure ^{235}U, then the oxygen in U_3O_8 is part of the matrix. Even the ^{238}U and other uranium isotopes are matrix material. The moisture and any other contaminants mixed with the U_3O_8 are matrix materials, as is even the container that holds the U_3O_8. If the can is inside a drum with fiberboard spacers, add the spacers to the matrix.

Some matrix materials are benign, but others affect the measured count rate in important ways. Anything with hydrogen can be important because neutrons lose much of their energy

when they scatter off hydrogen (think of the collision of two billiard balls). As hydrogen-bearing matrix is added near the fissile material, the count rate first goes up because the lower-energy neutrons induce more fissions. However, as more hydrogen is added, the count rate can go down as neutron captures in hydrogen compete with fissions. The impact varies with the position of the fissile material with respect to the hydrogenous matrix, making the count rate vary with position.[4]

A matrix of iron is much more benign because neutrons scatter without losing much energy (think of a ping-pong ball bouncing off a bowling ball). With 211 kg of iron in a 200-liter drum, the count rate was reduced by 4.5% compared with an empty drum,[4] a small but non-negligible fraction.

Techniques have been developed to mitigate the effect of hydrogen in 200-liter drums where the effects can be large simply because there is more room for matrix material than in small cans. The original technique used flux monitors built into the assay chamber. These were low-efficiency ^{3}He tubes whose outputs were counted during the irradiation. They counted a small fraction of the neutrons that came directly from the ^{252}Cf source, a larger fraction of neutrons that scattered off the moderating walls, and another large fraction of neutrons that entered the drum and returned with reduced energies. It is these last neutrons that we really want to count; the others are unavoidable background. One of the flux monitors was wrapped in cadmium to absorb low-energy neutrons, making it rather insensitive to the matrix in a drum. The other monitor rate changed significantly with the amount of hydrogen. The ratio of the flux monitors is independent of the ^{252}Cf source strength and can be used to correct for hydrogen.[4]

A hardware solution is to envelop the drum with about 1.9 cm of polyethylene. A sleeve made from a septic tank liner fits around a 200-liter drum very nicely; adding a top and bottom completes the envelopment. The result is a huge reduction in an otherwise large variation in count rate with the position of the fissile material. This happens because the first one or two collisions happen in the external polyethylene, reducing the range of energies of the neutrons entering the drum. The matrix effect is greatly reduced by the sleeve.[4] The calibration no longer has to deal with count rates changing with position.

However, a polyethylene sleeve increases the self-shielding problem because the neutron energies are lower. If the drum contains waste with small particle sizes, this is not an important issue. But if large pieces are present, the assay will be biased low by self-shielding.

If the positions of the uranium in a drum can be determined, a correction factor can be applied to each piece of uranium. This technique, developed for the standard 200-liter drum, does not require any extensive modification to the hardware. A stepping motor must be used for the turntable instead of an analog motor, but this is easy. The Compumotor Indexer Model 4000 can drive four motors, so even this module needs no change. The software needs extensive modification because the data collection and analysis is quite different from the conventional assay.

Six detector banks around the sides of a 200-liter drum, plus banks above and below the drum, are used to give a low-resolution (≈10 cm) “picture” of the distribution of fissile material within the drum. The drum does not rotate during a set of shuffles. The count in a bank depends on the relative positions of the ^{252}Cf source and the fissile material to that bank. To get enough information to locate the material, the drum is rotated 60° and another set of shuffles is done. This is continued for a full revolution of the drum. The mathematical algorithm is then applied to calculate the amount of fissile material in each of the 39 cells of equal volume within the drum. This algorithm uses calibration data from measurements on a standard placed sequentially in

each of the 39 (or so) cells. It is not necessary that the matrix be homogenous, as is usual with the conventional shuffler measurement, only that it is known and the calibration drum accurately reflects the distribution. The count time is approximately doubled with this technique, but this only means going from 15 min to 30 min.

For many matrices, there is no need to make a matrix correction. Cans of high-fired oxide have a rather benign and constant matrix (oxygen, steel walls) and what minor effects arise are automatically included in the calibration. Uranium carbide has an important matrix component, but it, too, is present in fixed relations with the uranium and can be included in the calibration as long as the geometric form is fixed. Each measurement should be analyzed for matrix effects and the most appropriate way to handle matrix problems.

B. Calculated Calibrations

What about materials that do not have calibration standards? The creation of new, certified standards is expensive and time consuming—a set of six U_3O_8 standards with 0.5 to 4.0 kg of uranium from the National Institute of Standards and Technology cost $100,000 in 1999. If the items of interest are unique, it makes little sense to create a standard and double the inventory. However, the calibration with U_3O_8 standards cannot be applied reasonably to U metal, UC_2, ^{233}U, ^{239}Pu, etc., because there will be large biases (Figure 13).

Can one calculate shuffler count rates and bypass the lack of physical standards? The topic is controversial, but it can be demonstrated to be very accurate. What is needed is not just an approximate calculation, but results that are as accurate as measured calibration data. This requires great attention to detail and as much benchmarking as possible. The close agreement between calculated and measured results is demonstrated in Figures 14 and 15. At the time of this writing, a methodology for accepting calculated calibrations for shufflers has been established at LANL.

A computational procedure for the standard 200-liter-drum shuffler has been developed[3] and can be readily applied to any other shuffler. For this large assay chamber, the ^{252}Cf source usually scans even a small can of oxide; scanning is not always necessary, but it is usually done. This means that the neutron flux entering the fissile material varies with time, greatly complicating the calculation. The situation would be much simpler if the source were stationary, so if the 200-liter-drum shuffler with a scanning source can be accurately described, the process will work for other shufflers.

Here are the steps in the procedure to calculate absolute shuffler count (MCNP stands for Monte Carlo Neutron and Photon code):

- Create an accurate MCNP model of the shuffler, including the fissile material and container.
- For each fissile mass, use MCNP to calculate the fission probabilities at representative positions of the ^{252}Cf source. For a 75-cm scan distance, a calculation at every 5 cm is adequate.
- Perform a separate MCNP calculation for the detection efficiency of delayed neutrons.
- Use the fission probabilities to calculate the six-group precursor populations at the end of the first irradiation. These are the numerical solutions of six differential equations.
- Calculate the count of delayed neutrons after the prescribed number of shuffles.
- Get the count rate by dividing the count by the sum of the individual count times.

- Use the set of masses and count rates to form the calibration curve.

This plan works and results in a reliable, accurate calibration curve. Ref. 3 provides details of the procedure and a comparison with measurements on uranium metal and oxides. A recently added delayed neutron patch in MCNPX simplifies the above process but both techniques work. Figure 14 shows measured and calculated rates for U_3O_8 calibration standards; there is no significant difference between the sets of calculated and the measured count rates. Similarly accurate calculations have been done for metals of uranium, plutonium, and their combination.

C. Enrichment Issues

The shuffler is used to measure the mass of ^{235}U, but ^{238}U is inevitably also present, and because it fissions, it adds to the delayed neutron count rate. If the enrichment is constant, the calibration curve automatically includes the effect and gives accurate ^{235}U masses (this is the case in Figure 14 with 92.4% enrichment).

If the enrichment is variable, two techniques have been applied to avoid biased results. The first is spectrum tailoring, which involves surrounding the ^{252}Cf source with a selection of metals to reduce the neutron energies below 1 MeV. This avoids fissioning the ^{238}U except by high-energy neutrons from fissions of ^{235}U; this effect is generally negligible. The Savannah River Uranium Scrap shuffler used spectrum tailoring successfully to avoid the enrichment problem.

Spectrum tailoring is not always practical. The flux of neutrons into the fissile material is reduced by the spectrum-tailoring materials, but a larger mass of ^{252}Cf can almost always be used to compensate. If the source scans a 200-liter drum, the spectrum-tailoring metal would have to extend over the full distance (75 cm), increasing the weight and cost accordingly. If a 550-μg source is normally used, the tailored source might need a mass of 1000 μg or more. The size of the shuffler would grow at least another 30 cm in all dimensions because of the additional shielding required.

The second technique is software based. If standards exist with different enrichments, the shuffler software can interpolate among calibration curves for different enrichments. The best approach to the enrichment issue at a particular facility needs to be decided early in the physics design phase by considering the measurement specifications, facility installation requirements, and the practicalities of shuffler designs.

V. DATA ANALYSIS

The assay chamber is the heart of the hardware and the data analysis is the heart of the software. Everything about a measurement comes together in the data analysis.

A. Raw Count Rates

Background counts are taken at the start of a measurement and delayed-neutron counts are taken after each irradiation. These are the “raw” counts that are converted into “raw” count rates using the measured times.

1. Background Counts

Before a background count is taken, the software ensures that the ^{252}Cf source is in the store position. Leakage of ^{252}Cf neutrons into the assay chamber increases rapidly as the source approaches the assay chamber, so it is important to put the source in a standard position where it contributes only a few counts a second. About a fourth of the assay time is spent on the background count. If the background is dramatically higher or lower than usual, the background count time should be changed to optimize the precision of the assay result.

2. Flux Monitor Counts

Flux monitors produce counts during the irradiations even if they are not used in the data analysis.

3. Post-irradiation Counts

These counts are from both background and delayed neutrons. None of these counts are corrected for dead-time losses because the rates are always low (e.g. under 10,000 cps).

B. Background Subtraction

The delayed neutron count rate is extracted from the post-irradiation count rate by subtracting the background count rate. The background rate for the inefficient flux monitors is always negligible compared to their very high count rates.

C. ^{252}Cf Decay Correction

The ^{252}Cf decays continuously and if this is not taken into account, measurements on the same item would be smaller each day. This is easily avoided by adjusting the delayed neutron count rate for the decay after some convenient reference date. The best value we have for the decay constant is 7.172×10^{-4} d^{-1}. A year after the reference date, the delayed neutron count rate is multiplied by 1.2995 to adjust for decay; after 5 years, the multiplier is 3.7062. The half-life of ^{252}Cf is known well enough to make this correction accurate for the useful life of the source.

No adjustment is included for ^{250}Cf neutrons because sources are not used more than 10 years and the number of ^{250}Cf neutrons is negligible compared with neutrons from ^{252}Cf, even after 20 years of decay (Figure 11).

D. Flux Monitor Matrix Correction

If hydrogenous matrices are involved in assays, the flux monitors might be used to correct for different amounts of hydrogen. The ratio of the flux monitor counts is a measure of hydrogen content, but the exact relationship depends on the nature of your materials and the shuffler design. A series of measurements on standard containers must be done to reveal the relationship and then built into the shuffler software.

Flux monitors are not needed if the matrices are not hydrogenous, if calibrations have been done for amounts of hydrogen that will be encountered, or if the position-correction style of assay is used.

E. Calibration Curve

The object of an assay is finally achieved when the corrected count rate *r* is used to calculate a mass of the fissile material *m* using a calibration curve. The only curve commonly used is a third-order polynomial with *r* expressed as a function *m* (as preferred by statisticians):

$$r = a_0 + a_1 m + a_2 m^2 + a_3 m^3.$$

Waste materials are likely to have only a_0 and a_1 nonzero. Measurements of oxides and metals up to 8 kg have been very well described by this polynomial. The four coefficients are determined from measurements on physical standards or from carefully calculated absolute count rates. They have a set of variances and covariances that can be specified and used to help calculate the uncertainty in a mass.

Figure 14 shows a calibration for U_3O_8 standards up to 8 kg. Self-shielding is evident for the first 750 g and then the curve becomes linear. The standards with masses of 3600 g or less are certified accurate; the higher masses are less certain.

The statistical uncertainties in the various counts are calculated and combined with the calibration variances and covariances to generate a total uncertainty (1σ) in the measured mass. In practice, the variances and covariances usually dominate the uncertainty[11], so careful and extensive measurements of calibration standards can have beneficial effects on the quality of future measurements.

F. Measurement Control

DOE regulations require a measurement control plan for an assay instrument. Periodic measurements on standards must be performed to check that the instrument is working properly. Each facility determines an appropriate plan, so only the basics are discussed here.

The standards chosen for measurement control checks might be calibration standards, but need not be. If oxide powders are chosen, the density of a powder is likely to change with handling and even without handling (thanks to gravity's relentless effort). Take measurements after shaking the can to loosen the powder and after tapping the can on a table top to settle the powder; the results may vary by 7% or more simply from a change of density that affects self-shielding. Using a powder for measurement control (or calibration) should include a handling procedure to ensure about the same density for all measurements. With this understood, powders have been used very successfully for measurement control purposes.

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